

LIDAR MEASUREMENTS FROM SPACE FOR TROPOSPHERIC CHEMISTRY INVESTIGATIONS:  
SUMMARY OF WORKSHOP OVERVIEW PRESENTATION

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## ABSTRACT

Over the past decade, NASA has played a lead role in defining the scientific objectives and technology requirements for spaceborne lidar investigations of the atmosphere. This paper presents an assessment of the potential for conducting lidar measurements from space for investigations that pertain specifically to tropospheric chemistry. In this paper, a description of potential lidar measurement techniques is given, and the scientific requirements for tropospheric chemistry are reviewed. The current status of airborne lidar measurements of aerosols,  $O_3$ , and  $H_2O$  is discussed, and a brief description of the evolution of lidar technology to space is given. Also, the measurement of tropospheric gases with a spaceborne lidar system is evaluated for a wide range of gas species. From this general assessment, it appears feasible to measure aerosols,  $H_2O$ ,  $O_3$ ,  $NH_3$ ,  $CO$ ,  $CH_4$ ,  $NO_2$ , atmospheric pressure and temperature, and winds with a lidar from space provided that the appropriate laser and receiver technology is available. For the mid-1990's, it is expected that lidar technology will be available for the measurement of aerosols,  $H_2O$ , and  $O_3$  from a space platform.

## INTRODUCTION

Lidar has been used extensively since the early 1960's for making measurements of various properties of the Earth's atmosphere. These include molecular and aerosol backscattering, gas concentration profiles, wind velocities, and atmospheric waves. The development of lidar in the early years was primarily concerned with laboratory and ground-based systems. Measurements with airborne lidar systems were not made until the early 1970's. By 1975, the lidar technique was well proven and consideration was being given to developing a shuttle-borne lidar system. A lidar system is now undergoing development to demonstrate the basic measurements of aerosol and cloud distributions from space.

There are four primary atmospheric lidar processes or techniques: elastic scattering, Raman scattering, resonance fluorescence, and differential absorption. Lidar measurements utilizing elastic backscattering have focused primarily on investigations of molecular density, aerosol and cloud distributions, and winds (e.g., Fiocco and Grams, 1966; Lane et al., 1971; Bilbro and Vaughan, 1978). Lidar systems detecting Raman-shifted frequencies have generally been limited to measurements of gases having high mixing ratios, such as water vapor, at relatively short ranges of 1 to 4 km at night (Cooney, 1971; Melfi and Whiteman, 1985). Resonance fluorescence has been an important technique used to measure sodium, potassium, and atmospheric waves in the upper atmosphere (e.g., Bowman et al., 1969; Megie et al., 1978; and Chanin and Hauchecorne, 1981). The Differential Absorption Lidar (DIAL) technique focuses on the simultaneous measurement of aerosol and gas concentration profiles, and it is the primary lidar technique for use in tropospheric gas measurements from space. This technique requires the near-simultaneous transmission of two wavelengths, usually by two tunable lasers. One of the lasers is tuned "on" the peak of an absorption line of the species to be measured, and the other is tuned "off" the absorption peak to a nearby wavelength. The two laser beams are backscattered from the

atmosphere to a collocated receiver where the signals are digitized as a function of range. The average gas concentration  $n$  between range  $R_1$  and  $R_2$  can be calculated using the expression (Schotland, 1974)

$$n = \frac{1}{2(R_2 - R_1)(\sigma_{\text{on}} - \sigma_{\text{off}})} \ln \frac{P_{\text{off}}(R_2) P_{\text{on}}(R_1)}{P_{\text{off}}(R_1) P_{\text{on}}(R_2)}$$

where  $\sigma_{\text{on}} - \sigma_{\text{off}}$  is the absorption cross section difference between the on and off wavelengths, and  $P_{\text{off}}(R)$  and  $P_{\text{on}}(R)$  are the detected backscattering signals received from range  $R$  for the on and off wavelengths, respectively.

As shown in figure 20, the development and application of lidar techniques have seen a steady growth over the last two decades. Early ground-based DIAL systems measured atmospheric  $\text{H}_2\text{O}$  and aerosols (Schotland, 1966; Browell et al., 1979);  $\text{SO}_2$  in power plant stack plumes (Hoell et al., 1975; Browell, 1982); and  $\text{O}_3$  in the lower atmosphere (Pelon and Megie, 1982). Recently, an advanced airborne DIAL system was developed and has been used to measure  $\text{O}_3$ ,  $\text{H}_2\text{O}$ , and aerosols in the troposphere (Browell et al., 1983). Studies of spaceborne lidar measurements and systems can be traced back to the early 1970's. The most comprehensive study of the science objectives and measurement capabilities of a spaceborne lidar system was published by NASA in 1979 (NASA, 1979). These activities led to the development of an autonomous DIAL system for the measurement of  $\text{H}_2\text{O}$  and aerosols from a high-altitude ER-2 aircraft as a precursor to a spaceborne DIAL system.

A working group was formed by NASA in 1978 to identify the major measurement requirements in tropospheric chemistry and to develop the scientific rationale and recommended research activities to be conducted by NASA to increase our understanding of the troposphere (NASA, 1981). A summary of the measurement requirements identified by that group is given in table X. A minimum of two gas concentration measurements in the troposphere, one across the boundary layer and one across the free troposphere, was determined to be necessary for most tropospheric chemistry applications. Listed in table X are the minimum detectable gas concentration levels that would be required. The measurement of atmospheric state variables was not identified to be of strong importance in tropospheric chemistry applications, and thus, relatively low accuracies were needed for the measurement of these parameters. Information on aerosols was cited as important for the understanding of heterogeneous chemistry, atmospheric structure, and atmospheric transport.

This paper discusses the status of current airborne lidar measurements of aerosols,  $\text{O}_3$ , and  $\text{H}_2\text{O}$  and the planned evolution of lidar technology to space. A general evaluation of gas measurements with lidar from space is presented. Future developments in lidar techniques and measurements that are suitable for space applications are assessed, and technology requirements for future spaceborne lidar systems for tropospheric measurements are presented.

#### AIRBORNE LIDAR MEASUREMENTS OF AEROSOLS, $\text{O}_3$ AND $\text{H}_2\text{O}$

An advanced airborne DIAL system has been developed at the NASA Langley Research Center for the investigation of aerosols,  $\text{O}_3$ , and  $\text{H}_2\text{O}$  in the troposphere (Browell et al., 1983). A schematic of the airborne DIAL system mounted in the NASA Wallops

Electra aircraft is shown in figure 21. In this system, two frequency doubled Nd:YAG lasers are used to pump two high conversion efficiency dye lasers. The on and off wavelengths are produced in sequential laser pulses with 100 to 250  $\mu$ s separation. Backscattered lidar returns at the two wavelengths are sequentially detected by a photomultiplier tube, digitized, and stored on high-speed magnetic tape. Gas concentrations and aerosol backscattering profiles are calculated for each measurement in real time by a minicomputer. The selection of the lasers was dictated by three basic requirements: tunability, high power, and reliability. The dye laser is easily tuned with different dyes over a wide selection of wavelengths from 400 to 1200 nm, and the output can be frequency doubled into the UV. The performance parameters of the airborne DIAL system operating in the UV and near-IR are listed in table XI.

Examples of airborne DIAL data are shown in figure 22 to illustrate some of the capabilities of the system. Signal returns from the UV (300 nm) and visible (600 nm) channels are displayed at the top of the figure to show the relative contrast resulting from increased aerosol backscattering in the atmospheric boundary layer. The range-corrected visible channel is shown at the lower left, and a gray scale picture made using these data is shown at the lower right. Each vertical line of the gray scale represents a laser shot, and the darker regions represent greater atmospheric backscattering, or aerosol concentration. This gray scale clearly defines the boundary layer height and presence of clouds (dark regions followed by absence of return signal from lower altitudes). This type of aerosol data can provide information on widely varying atmospheric conditions including aerosol layering and transport, clouds, condensation levels, and topographic features. Figure 23 presents an intercomparison of DIAL  $O_3$  measurements with in situ measurements made onboard a small aircraft spiraling in the vicinity of the Electra. The intercomparison shows that: (1) the remote lidar measurements and in situ measurements are in excellent agreement, (2) the DIAL measurements have 10 percent or less measurement uncertainty for the spatial resolution shown, and (3) the DIAL measurements can be made with high vertical (210 m) and horizontal (6 km) resolution.

Airborne DIAL measurements have been used to study the correlation between spatial distributions of  $O_3$  and aerosols in the troposphere and lower stratosphere. Figure 24 shows  $O_3$  measurements by the DIAL system in the zenith-viewing mode compared with in situ observations. Profiles of potential temperature and ozone measured by an ozonesonde are presented in the figure along with DIAL  $O_3$  data. The location of a change in slope of the potential temperature profile ( $\approx 11$  km) defines the height of the tropopause. The  $O_3$  profiles show layering of  $O_3$  in the lower stratospheric regions, and there is good agreement between the DIAL and in situ measurements. Further, other DIAL measurements show that this  $O_3$  layering persisted along the flight path of the aircraft. This indicates the ability of an airborne DIAL system to map  $O_3$  layers in the troposphere and lower stratosphere and to study stratosphere and troposphere exchanges.

When the airborne DIAL system is used to study  $H_2O$  profiles, the on-line laser wavelength is tuned to coincide with the peak of a  $H_2O$  absorption line in the 720 nm region. An intercomparison between the DIAL  $H_2O$  measurements and in situ observations is shown in figure 25. The good comparison between these two observations shows the ability of the DIAL system to make  $H_2O$  measurements in the lower troposphere. This capability was used for the first time in the fall of 1982 for the study of marine boundary layer characteristics in the vicinity of the Gulf Stream Wall (Browell et al., 1984).

## EVOLUTION OF LIDAR TECHNOLOGY FOR SPACE

There are three lidar development programs within NASA that are directly aimed at providing the technological base for placing a DIAL system on a free-flying space platform by the mid 1990's. The LASE (Lidar Atmospheric Sensing Experiment) and LITE (Lidar In-Space Technology Experiment) projects address important phases of the spaceborne lidar development program. The LASA (Lidar Atmospheric Sounder and Altimeter) system is the near-term goal for a lidar system operating on a polar orbiting platform. This would be part of the NASA EOS Program.

The LASE system is being developed as a high altitude (16-21 km), autonomous DIAL system for operation on a NASA ER-2 (Extended Range U-2) aircraft. It will initially operate in the 727-nm wavelength region to measure H<sub>2</sub>O profiles in the lower troposphere, and at a later stage it will be used in the 940-nm wavelength region to measure H<sub>2</sub>O profiles in the upper troposphere. In addition, atmospheric pressure and temperature determinations will be made using DIAL O<sub>2</sub> measurements in the 760-nm wavelength region. Figure 26 shows the results of our simulations to determine the random errors associated with H<sub>2</sub>O measurements with the LASE system. The random error at a specific altitude depends upon the absorption strength of the H<sub>2</sub>O line being used, with the stronger lines being more suitable for higher altitude regions. Clearly the random errors for night operation and for a horizontal resolution of 20 km and a vertical resolution of 200 m would permit a 5- to 10-percent measurement uncertainty in a specific altitude region. Our analysis shows that systematic errors could contribute a significant amount to the uncertainty of these measurements. The systematic errors can be caused by: (1) interaction of the laser linewidth with the altitude dependent H<sub>2</sub>O absorption linewidth, (2) Doppler broadening of backscattered light by air molecules, (3) accuracy in the positioning of the laser line with respect to the H<sub>2</sub>O line, (4) uncertainty in knowledge of the laser wavelength, and (5) the broadband laser energy component outside the dominant laser on-line emission. Our simulations indicate that with proper controls on the laser output and by folding in model atmospheric temperature and pressure information, the systematic errors could be limited to a few percent (Ismail et al., 1984). The LASE system would be capable of making measurements on long-range flights (>4500 km) during both day and night background conditions, and because of its unique autonomous mode of operation, the LASE system would be a precursor to the development of a spaceborne lidar system. LASE is expected to make its initial flight in late 1988.

The development of spaceborne lidar systems is contingent on the ability of lasers to operate in a space environment. NASA is taking a step in the direction of developing an operational laser capability for space with the LITE project. The objective of LITE is to develop the technology base and measurement techniques necessary to operate a solid-state laser in a spaceborne lidar system. The approach being used is to space-harden existing lidar technology with minimum change. The initial experiment will utilize a frequency doubled and tripled Nd:YAG laser in a lidar system with simultaneous measurements at three wavelengths. The construction will incorporate easily modified modular concepts, and the experiments will focus on aerosol, cloud, and atmospheric density measurements. LITE is scheduled to fly on the Space Shuttle in 1989.

The next program following LITE to incorporate a lidar system in space will most likely be the EOS Program scheduled for the middle 1990's time period. A lidar system for EOS is being proposed, and it is called the LASA instrument. The science objectives for LASA cover many disciplines, including hydrology, altimetry, geodynamics, aerosol, and cloud studies. The list of lidar measurements proposed for LASA is

shown in table XII. The 14 listed items have all been demonstrated in the laboratory, and most have been demonstrated in both ground-based and low-altitude airborne systems. LASE data from the ER-2 aircraft will further demonstrate high-altitude lidar measurement capabilities. Using the technology base developed from the LASE and LITE programs, LASA should be successful in gathering large quantities of data on a global basis to make a significant step forward in the scientific understanding of our atmosphere.

#### GAS MEASUREMENTS WITH LIDAR FROM SPACE

The accuracy of a gas concentration measurement with the DIAL technique is dependent upon the absorber thickness across the region where the average gas concentration is to be determined. Figure 27 shows the sensitivity in the error in optical depth determination to the absorber optical depth for various transmittance measurement errors. For small absorber optical depths, the error in the determination of the optical depth decreases with increasing absorber optical depth. An optimum absorber optical depth is reached around a value of 1 (note that this analysis is for a single-pass measurement), and at higher optical depths the error begins to increase precipitously due to loss of signal. Thus, the measurement of a gas using the DIAL technique depends upon the expected gas concentration, the available gas absorption cross sections, and the range over which the absorption is to be measured.

Table XIII lists the mean mixing ratios for the tropospheric gases identified in Table X. Also given in table XIII are the column burdens for the same gases over the entire troposphere. To evaluate the ability of a DIAL system to make a measurement of a gas from space, we must next look at the available absorption cross sections for these gases. In table XIV, the gases from table XIII are listed in order of decreasing mixing ratio in the lower troposphere. For each molecule, the absorption wavelength region is given with an estimate of the maximum absorption cross section for that region (weak lines are not usually difficult to find in the same region). Since the DIAL measurement actually involves a double pass across any altitude segment, estimates of the two-way optical are given across a 1-km boundary layer and across the entire troposphere.

From the information provided in table XIV and from the results of other spaceborne lidar experiment simulations not presented here, the following measurements have been identified as potential objectives for a spaceborne DIAL system, providing the laser and receiver technology are available: (1) profile measurements of  $H_2O$ ,  $O_3$ ,  $CO$ ,  $CH_4$ , and possibly  $NH_3$ ; (2) tropospheric column content measurements of  $NO_2$  and possibly  $NO$  and  $SO_2$  with very low-horizontal resolution; (3) atmospheric state parameter measurements to the required accuracy and direct measurements of winds; and (4) aerosol distribution measurements for information on atmospheric structure and meteorological parameters, and multiple-wavelength aerosol measurements for information on aerosol composition and mass loading.

An example is given in figure 28 of the accuracy for obtaining  $H_2O$  profile measurements using absorption lines of different strengths in the 727 nm wavelength region. A vertical resolution of 1.0 km and a horizontal resolution of 100 km were used in these calculations with a laser energy of 500 mJ at 10 Hz and a 1.25-m diameter telescope receiver system. It can be seen that the weaker lines optimize at a lower altitude, and that to provide altitude coverage from the surface to the upper troposphere, the DIAL measurement requires the use of at least two absorption lines.

With the resolution shown, the H<sub>2</sub>O profiles measurement could have a random error of <10% below 5 km altitude and <15% from 3-10 km altitude. These measurement uncertainties can be further reduced by using a larger vertical and/or horizontal resolution.

## STATUS OF DIAL MEASUREMENTS SUITABLE FOR SPACE APPLICATIONS

Table XV summarizes the status of DIAL measurements that pertain to eventual remote sensing of tropospheric parameters from space. The gas species or atmospheric parameter is listed along with the nominal wavelength region for the lidar measurement. In all cases, the theoretical feasibility for an atmospheric lidar measurement has been shown. The atmospheric measurements from a laboratory environment have also been demonstrated from most of the parameters. Some of measurements were made in enhanced gas concentrations in plumes, in an urban environment, or over a roadway. The others were made in the ambient rural or remote troposphere. Fewer field measurements have been made with mobile or portable ground-based lidar systems. Most of these systems have been used for pollution studies of SO<sub>2</sub> or in tracking plumes using aerosols as a tracer. Lidar measurements from aircraft are indicated along with the year the first demonstration was conducted. As can be seen from table XV, all of the activity in airborne DIAL measurements has been aimed at O<sub>3</sub> and H<sub>2</sub>O over the last 6 years. Table XVI presents an estimate of where airborne DIAL measurements are going in the near future (<2 years) and beyond. Also indicated is the type of laser or wavelength region that is expected to be used in the measurement. In the near future O<sub>3</sub> will be measured with an excimer-based DIAL system as will SO<sub>2</sub> in power plant plumes. Atmospheric H<sub>2</sub>O, pressure, and temperature will be measured using Alexandrite lasers in an airborne DIAL system. Beyond the next two years, DIAL measurements of CH<sub>4</sub>, CO, NH<sub>3</sub>, NO<sub>2</sub>, and NO depend upon the development of appropriate lasers and DIAL systems in the wavelength regions shown.

The general characteristics needed for spaceborne lasers and a list of potential tunable lasers for spaceborne lidar applications are given in table XVII. Most spaceborne lidar applications require tunable laser wavelengths with high average power. The characteristics given in table XVII are for a free-flying satellite that may only be visited every 12 to 24 months. The types of tunable lasers given in the table are examples of general laser types that are currently under development and are potential candidates for future spaceborne lidar missions.

## SUMMARY

This paper has discussed the evolution of lidar systems for tropospheric observations. Measurements of aerosols, O<sub>3</sub>, and H<sub>2</sub>O made with the NASA LaRC airborne DIAL system were presented as examples of the types of data that could be obtained from a spaceborne lidar system. An autonomous DIAL system under development by NASA for the high-altitude ER-2 aircraft was described. Future spaceborne lidar systems, LITE and LASA, were discussed, as well as the requirements for future spaceborne laser systems. It is clear that the successful development of spaceborne lidar systems depends critically on solving the laser technology challenges of improved laser efficiency and long lifetimes.

Spaceborne lidar can potentially contribute to tropospheric chemistry investigations by making profile measurements of H<sub>2</sub>O, O<sub>3</sub>, CO, CH<sub>4</sub>, and possibly NH<sub>3</sub>. Also, it may be possible to measure the tropospheric column content of NO<sub>2</sub> and possibly SO<sub>2</sub>

with low horizontal resolution. Winds and atmospheric state variables could also be provided by lidar measurements. Aerosol and cloud characterization can be obtained with a multiple-wavelength lidar system. All of these measurements require the appropriate laser and receiver technology for long-term space applications. By the mid-1990's a spaceborne DIAL system could be placed in orbit to investigate tropospheric profiles of aerosols and  $H_2O$ , or possibly investigate low vertical resolution ( $>3$  km) measurements of tropospheric  $O_3$ . The information obtained by these first systems would make an important contribution to tropospheric chemistry investigations. By the year 2000, laser and lidar technology will have advanced to the point where the remote measurement of many other species would be possible. Lidar is an important area of development because there are no other remote sensing techniques that can obtain high vertical resolution ( $<2$  km) measurements of atmospheric parameters and gases in the lower atmosphere from space.

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TABLE X.- PRELIMINARY MEASUREMENT REQUIREMENTS IN TROPOSPHERIC CHEMISTRY\*

Resolution - Vertical: Boundary Layer & Free Trop. Minimum  
Horizontal:  $\leq 200$  km

Species (Accept. Detect.) -  $O_3$  (10 PPB), CO (50 ppb),  $CH_4$  (1.5 ppm) NMHC (10 ppt),  
 $H_2CO$  (0.10 ppb), NO (10 ppt),  $NO_2$  (10 ppt),  
 $HNO_3$  (10 ppt), OH ( $5 \times 10^5 \text{ cm}^{-3}$ ),  $HO_2$  ( $10^8 \text{ cm}^{-3}$ ),  
 $H_2O_2$  (1.0 ppb),  $H_2O$  (100 ppm),  $NH_3$  (0.10 ppb),  
 $SO_2$  (50 ppt),  $H_2S$  (5 ppt),  $CH_3SCH_3$  (5 ppt),  $CS_2$  (5 ppt),  
COS (50 ppt)

State Variables - Temp. ( $\pm 5K$ ), Press. ( $\pm 10\%$ ), Cloud Cover and Height, Freq. and  
Dist. of Lightning

Aerosol Measurements - Optical Properties Under Flight Track, Chemical Composition,  
Size Distribution

\*Adapted from NASA, 1981

TABLE XI.- AIRBORNE DIAL SYSTEM CHARACTERISTICS

Transmitter:

Two Pump Lasers -- Quantel Model 482

Pulse Separation -- 100  $\mu s$

Pulse Energy -- 350 mJ at 532 nm

Repetition Rate -- 10 Hz

Pulse Length -- 15 ns

Two Dye Lasers -- Jobin Yvon Model HP-HR

	UV (near 300 nm)	Near-IR (near 720 nm)
Fundamental Dye Output Energy	157 mJ pulse near 600 nm	63 mJ pulse
Doubled Dye Output Energy	47 mJ/pulse near 300 nm	--
Transmitted Laser Energy	40 mJ/pulse near 300 nm and 80 mJ/pulse near 600 nm	50 mJ/pulse
Laser Linewidth	<4 pm	<2 pm

Receiver:

Area of Receiver	0.086 m <sup>2</sup>	0.086 m <sup>2</sup>
Receiver Efficiency to PMT	28%	29%
PMT Quantum Efficiency	29%	4.8%
Total Receiver Efficiency	8.1%	1.4%
Receiver Field of View	2 mrad	2 mrad

TABLE XII.- DEMONSTRATED LIDAR MEASUREMENTS FOR LASA

	Lab	Ground-Based	Low Alt. Airborne	High Alt. Airborne	LASA Sci. Objectives
1. Altimetry	X	X	X	-	2
2. Retro-Ranging	X	X	-	-	3
3. Cloud Top Height	X	X	X	X	1
4. PBL* Height	X	X	X	X	1
5. Strat. Aero.	X	X	X	-	4
6. Cloud Prop.	X	X	X	X	4
7. Trop. Aero.	X	X	X	X	4
8. H <sub>2</sub> O Column	X	X	X	A	1
9. Surface Pres.	X	-	-	-	5
10. O <sub>3</sub> Column	X	X	X	-	5
11. H <sub>2</sub> O Profile	X	X	X	A	1
12. Pres. Profile	X	X	-	-	5
13. Temp. Profile	X	-	-	-	5
14. O <sub>3</sub> Profile	X	X	X	-	5

LASA Science Objectives: 1 Hydrology; 2 Altimetry; 3 Geodynamics;  
4 Aerosols and Clouds; 5 Other

A: LASE Measurements (1988)

\*Planetary Boundary Layer

TABLE XIII.- TROPOSPHERIC GAS ABUNDANCES

Species	Mean Mixing Ratio in Lower Trop.*	Nominal Tropospheric Column Burden** (ppm-m)
O <sub>3</sub>	50 ppb	230
CO	90 ppb	800
CH <sub>4</sub>	1.65 ppm	1.1 x 10 <sup>4</sup>
NMHC* (C <sub>2</sub> to C <sub>12</sub> )	> 1 ppb	-
H <sub>2</sub> CO	-	0.14
NO	5 ppt	2.0
NO <sub>2</sub>	10 ppt	1.7
HNO <sub>3</sub>	50 ppt	23
OH	0.05 ppt	0.01
HO <sub>2</sub>	1 ppt	0.12
H <sub>2</sub> O <sub>2</sub>	500 ppt	-
H <sub>2</sub> O	1300 ppm	4.1 x 10 <sup>7</sup>
NH <sub>3</sub>	10 ppb	20
SO <sub>2</sub>	100 ppt	0.81

\*NASA, 1981

\*\*Alvarado et al., 1982

TABLE XIV.- TROPOSPHERIC GAS ABSORPTION PROPERTIES

Species	Wavelength Region ( $\mu\text{m}$ )	Absorption Cross Section ( $\text{cm}^2/\text{Molecule}$ )	Est. of Maximum Two-Way Optical Depth	
			Across 1 km B.L.	Across Troposphere
$\text{H}_2\text{O}$	0.72	$6 \times 10^{-23}$	0.42	13
	0.94	$2 \times 10^{-21}$	14	---
	1.23	$4 \times 10^{-20}$	280	---
	6.3, 2.7	$1 \times 10^{-18}$	7000	---
$\text{CH}_4$	7.7, 3.33, 3.25	$1 \times 10^{-18}$	8.9	---
$\text{CO}$	4.6	$2 \times 10^{-18}$	0.97	8.6
	2.33	$2 \times 10^{-20}$	0.010	0.086
$\text{O}_3$	10	$1 \times 10^{-19}$	0.027	0.12
	0.29	$9 \times 10^{-19}$	0.24	1.07
$\text{NH}_3$	10	$1 \times 10^{-18}$	0.05	0.1
$\text{SO}_2$	4.0	$3 \times 10^{-20}$	$1.6 \times 10^{-5}$	$1.3 \times 10^{-4}$
	0.3	$1 \times 10^{-18}$	$5.4 \times 10^{-4}$	.0044
$\text{NO}_2$	0.44	$3 \times 10^{-19}$ ( $\Delta\sigma$ )	$1.6 \times 10^{-5}$	0.0028
$\text{NO}$	5.4, 5.2	$1 \times 10^{-18}$	$2.7 \times 10^{-5}$	0.011

TABLE XV.- STATUS OF DIAL MEASUREMENTS SUITABLE FOR SPACE APPLICATIONS

Gas Species/ Atm. Param.	Nominal Wavelength Range ( $\mu\text{m}$ )	Theoretical Feasibility	Atm. Mea. from Lab	Field Mea.	A/C Mea.
$\text{O}_3$	0.3	X (RR)	X (RR)	X (RR)	1980 (RR)
	9.5	X (CC & RR)	X (CC)	X (CC)	1979 (CC)
$\text{H}_2\text{O}$	0.72	X (RR)	X (RR)	X (RR)	1982 (RR)
	0.94/1.23	X (RR)	--	--	
	1.8	X (CC)	X (CC)	--	
	10.3	X (CC & RR)	X (CC & RR)	X (CC)	1982 (CC)
$\text{CH}_4$	3.5	X (CC)	X (CC)	--	
$\text{CO}$	5.0	X (CC)	X (CC-Roadway)	--	
$\text{NH}_3$	10.3	X (CC)	X (CC)	--	
$\text{SO}_2$	0.3	X (RR)	X (RR-Plume)	X (RR-Plume)	
$\text{NO}_2$	0.44	X (RR)	X (RR-Plume,	X	
	--	--	CC-Urban)	--	
$\text{NO}$	5.3	X (CC)	X (CC-Roadway)	--	
Press. ( $\text{O}_2$ )	0.76	X (RR)	X (RR)	X	
Temp. ( $\text{O}_2$ )	0.78	X (RR)	X (CC)	X	
Aerosol Dist.	0.3/0.6/1.0	X	X	X	1968
Aerosol Char.	0.3/0.6/1.0	X	X	X	1984
Winds (Doppler)	10.6	X (Profile)	X	X	1981

TABLE XVI.- AIRBORNE LIDAR TROPOSPHERIC MEASUREMENTS

<u>Demonstrated</u>	<u>Near Future (<math>\leq</math> 2 Years)</u>	<u>Future (<math>\geq</math> 2 Years)</u>
O <sub>3</sub> (Nd:YAG-DYE; CW-CO <sub>2</sub> )	O <sub>3</sub> (Excimer)	O <sub>3</sub> (Pulsed CO <sub>2</sub> )
H <sub>2</sub> O (Nd:YAG-DYE; CW & Pulsed CO <sub>2</sub> )	H <sub>2</sub> O (Alex.)	H <sub>2</sub> O (0.94/1.23 $\mu$ m)
SF <sub>6</sub> Tracer (CW-CO <sub>2</sub> )	SO <sub>2</sub> (Excimer)	CH <sub>4</sub> (3.3 $\mu$ m)
Aerosol & Cloud Dist. (Nd:YAG)	P (Alex.)	CO (4.6 $\mu$ m)
Aerosol Discrim. (N $\lambda$ Nd:YAG-DYE)	T (Alex.)	NH <sub>3</sub> (10.3 $\mu$ m)
P (2 $\lambda$ Nd:YAG)	--	NO <sub>2</sub> (0.44 $\mu$ m)
Winds (CW & Pulsed-CO <sub>2</sub> Doppler)	--	NO (5.4 $\mu$ m)

TABLE XVII.- SPACEBORNE LASERS

<u>General Characteristics</u>	<u>Tunable Laser Types</u>
High Power (1 - 20 W)	Alexandrite (720 - 770 nm)
Wavelength Tunable (10 - 40 cm <sup>-1</sup> )	Co:MgF <sub>2</sub> * 1.6 - 2.3 $\mu$ m
Narrow Linewidth (<0.02 cm <sup>-1</sup> )	Ti:Sapphire (700 - 850 nm)
Long Lifetime (12 - 24 mo)	Emerald (751 - 759 nm)
Ruggedized (Shuttle Launch; Space Environ.)	Nd:Glass*
Reduced Complexity and Modular	Excimer*
Capable of Meeting Eye Safety Criteria	CO <sub>2</sub> (Line Tunable and Isotope Broadened)

\*Raman Shifted

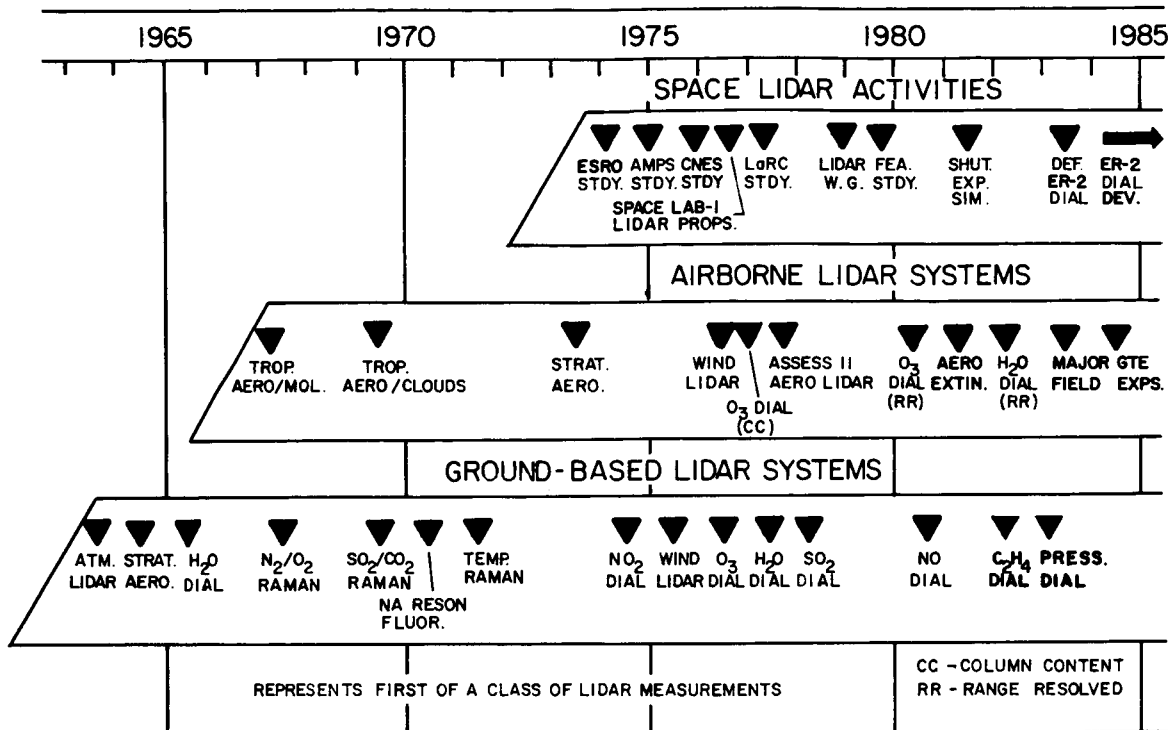


Figure 20.- History of lidar development with each milestone representing the first in a class of lidar measurements. In the case of ground-based and airborne lidar measurements the measurement parameter/species are given along with the lidar technique used. Various studies are cited in the activities supporting spaceborne lidar system development.

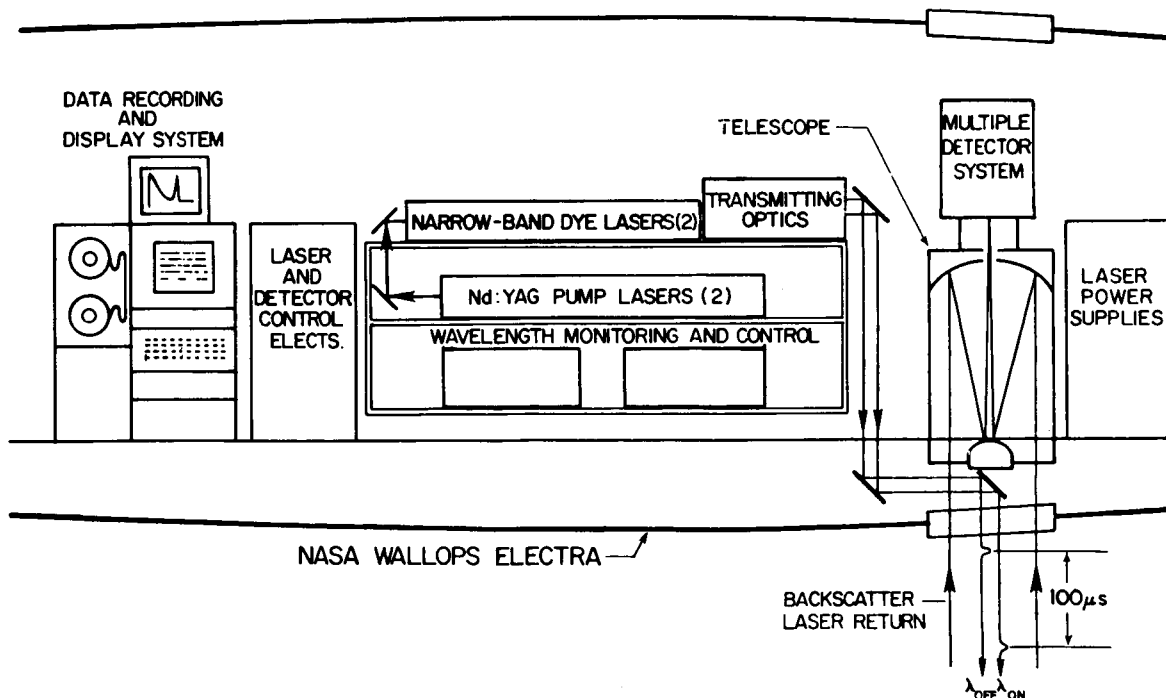


Figure 21.- NASA Langley Research Center airborne DIAL system schematic (Browell et al., 1983).

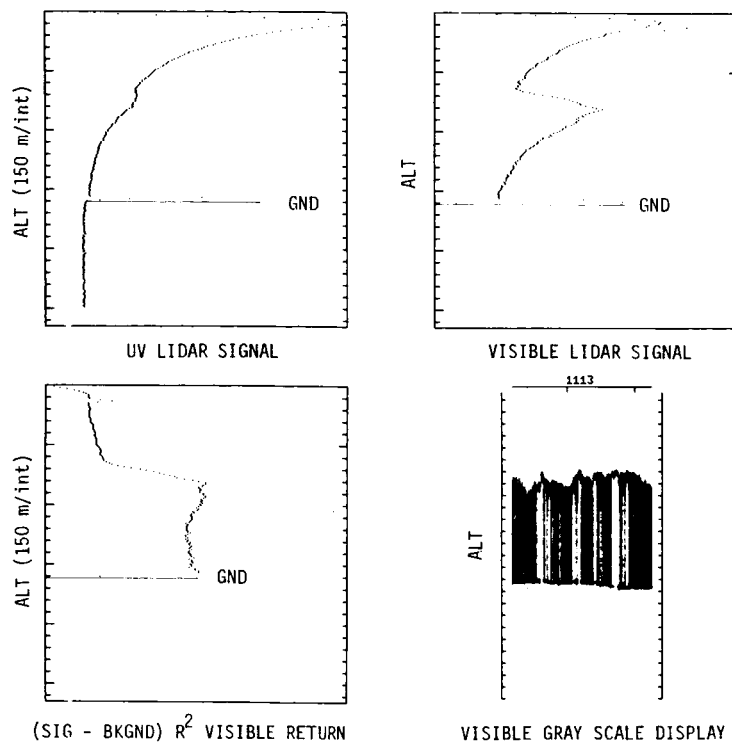


Figure 22.- Airborne DIAL UV and visible lidar returns with DIAL system operating in a nadir mode from an aircraft altitude of 3 km.

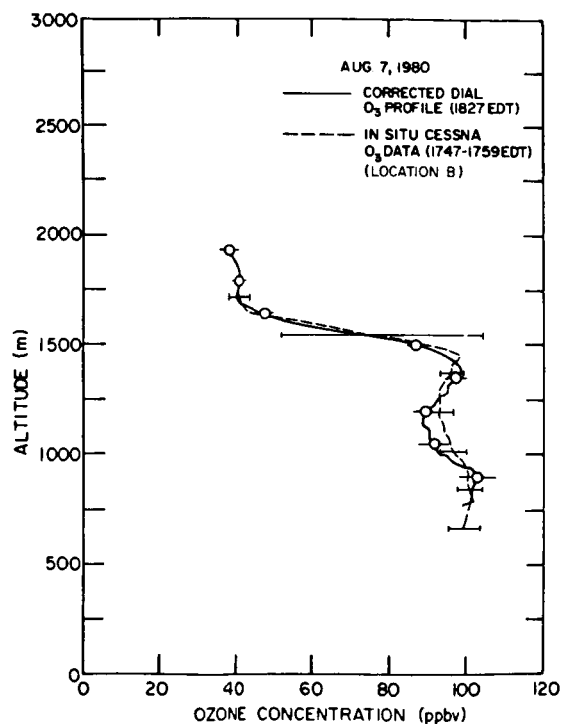


Figure 23.- Comparison of airborne DIAL and in situ measurements of O<sub>3</sub> profiles (Browell et al., 1985).

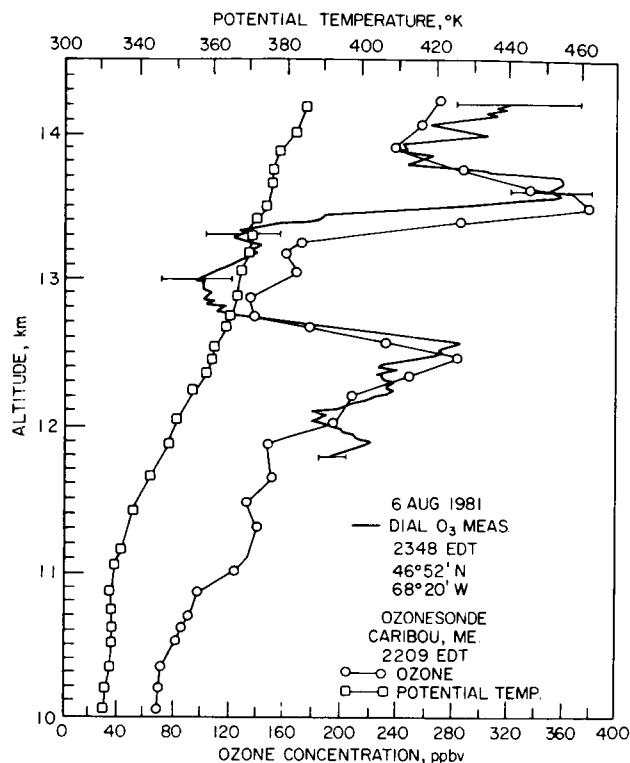


Figure 24.- Airborne DIAL and in situ  $O_3$  measurements in the lower stratosphere (Browell, 1983).

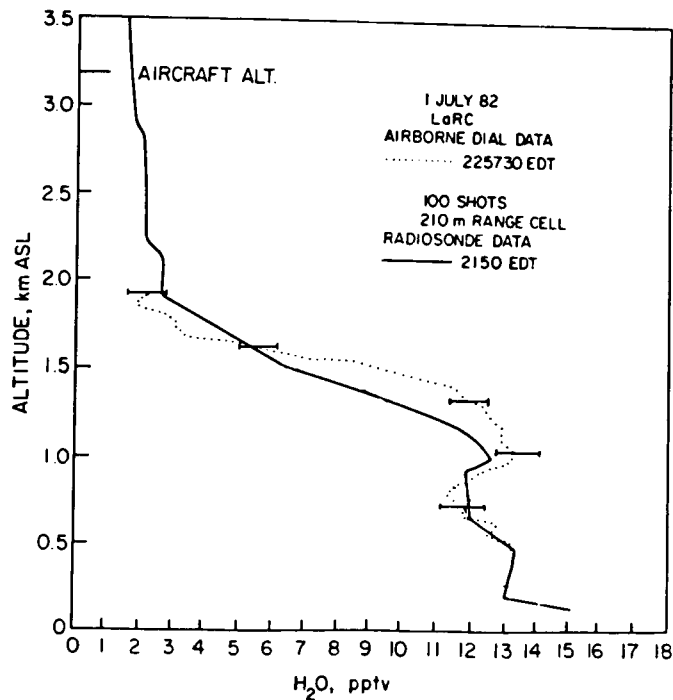


Figure 25.- Comparison of airborne DIAL and radiosonde measurements of  $H_2O$  profiles.

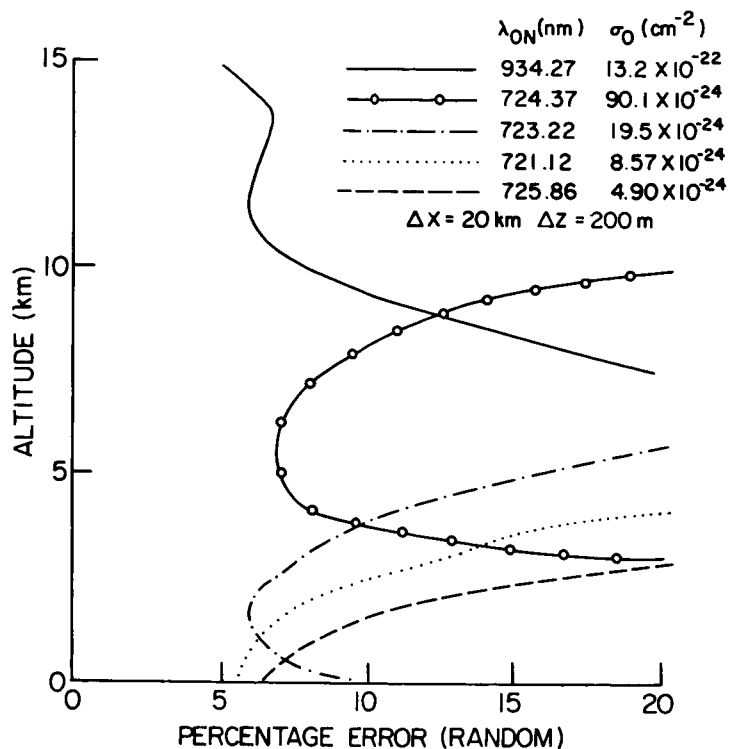


Figure 26.- Simulations of H<sub>2</sub>O measurement uncertainties for the ER-2 DIAL system. An aircraft altitude of 16 km and a mid-latitude summer H<sub>2</sub>O profile was assumed.

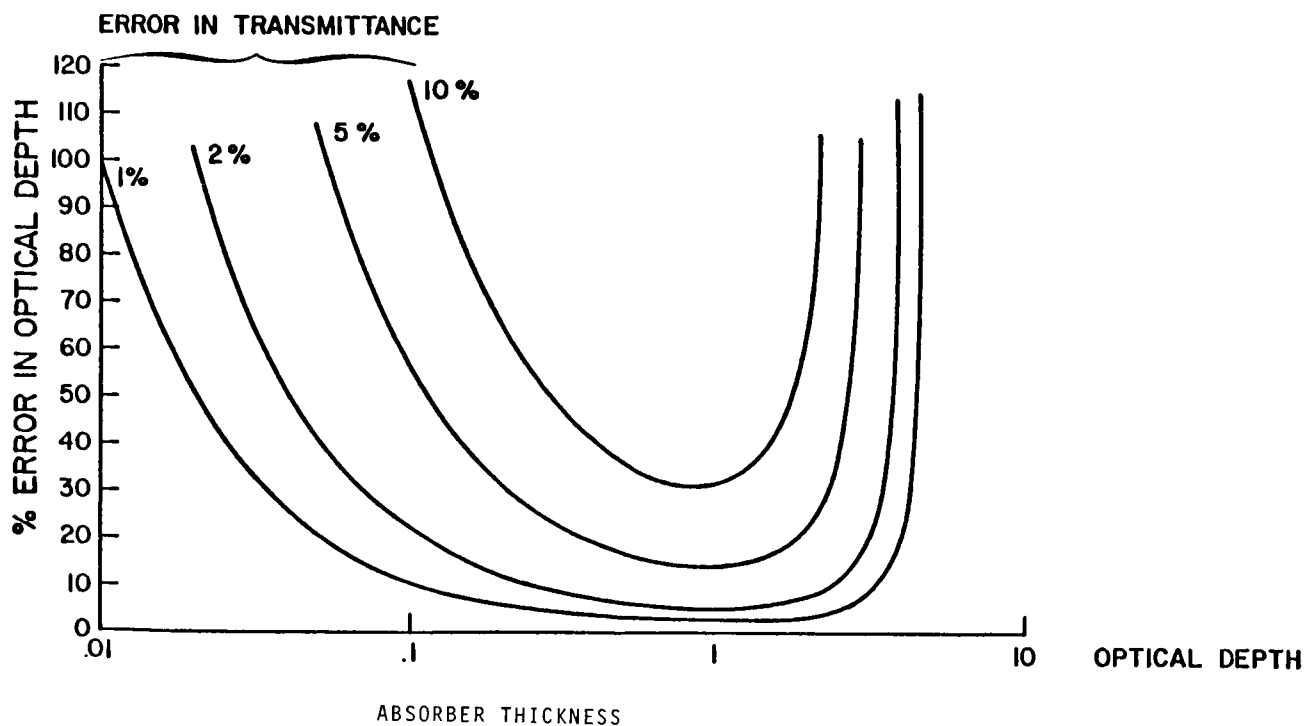


Figure 27.- Dependence of optical depth measurement error on absorber optical depth and transmittance measurement accuracy.



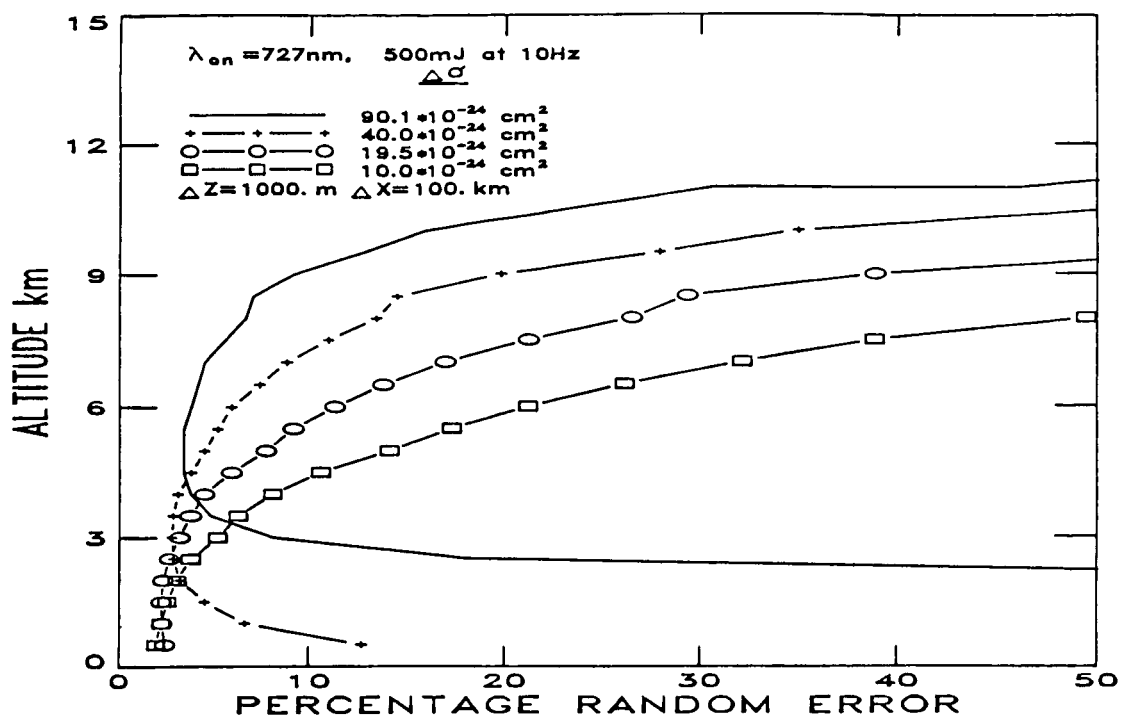


Figure 28.- Spaceborne DIAL H<sub>2</sub>O profile measurement uncertainties. An orbital altitude of 800 km and a mid-latitude summer H<sub>2</sub>O model profile were assumed.

# APPENDIX A: TROPOSPHERIC CHEMISTRY: AN OVERVIEW

Joel S. Levine

## OXYGEN SPECIES

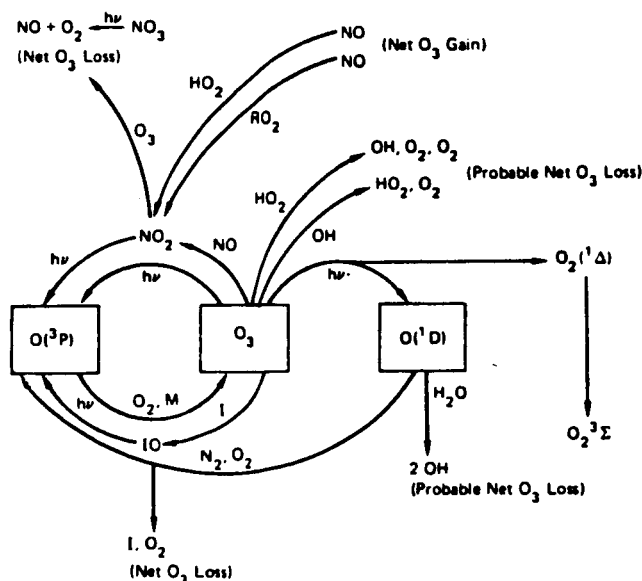
SPECIES	CONCENTRATION <sup>1</sup>	LIFETIME <sup>2</sup>	COMMENTS <sup>3</sup>
OZONE (O <sub>3</sub> )	10 - 100 PPBV	WEEKS TO MONTHS	C, TRANSPORT FROM STRATOSPHERE; GREENHOUSE SPECIES
ATOMIC OXYGEN (O)	10 <sup>3</sup> CM <sup>-3</sup>	V.S.	C, LEADS TO O <sub>3</sub> PRODUCTION
EXCITED ATOMIC OXYGEN (O( <sup>1</sup> D))	10 <sup>-2</sup> CM <sup>-3</sup>	V.S.	C, LEADS TO OH PRODUCTION

<sup>1</sup>LEVINE, J.S. (EDITOR), 1985: THE PHOTOCHEMISTRY OF ATMOSPHERES: EARTH, THE OTHER PLANETS, AND COMETS, ACADEMIC PRESS, INC., PP. 6-7.

<sup>2</sup>IBID, P. 44: V.S. = VERY SHORT (SECONDS OR LESS); V.L. = VERY LONG (DECADES OR LONGER)

<sup>3</sup>A = ANTHROPOGENIC; B = BIOGENIC; C = CHEMICAL OR PHOTOCHEMICAL

## OXYGEN SPECIES CHEMISTRY



Photochemical production and destruction of O<sub>3</sub>. Key oxygen species are shown in solid boxes.

FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.

## HYDROGEN SPECIES

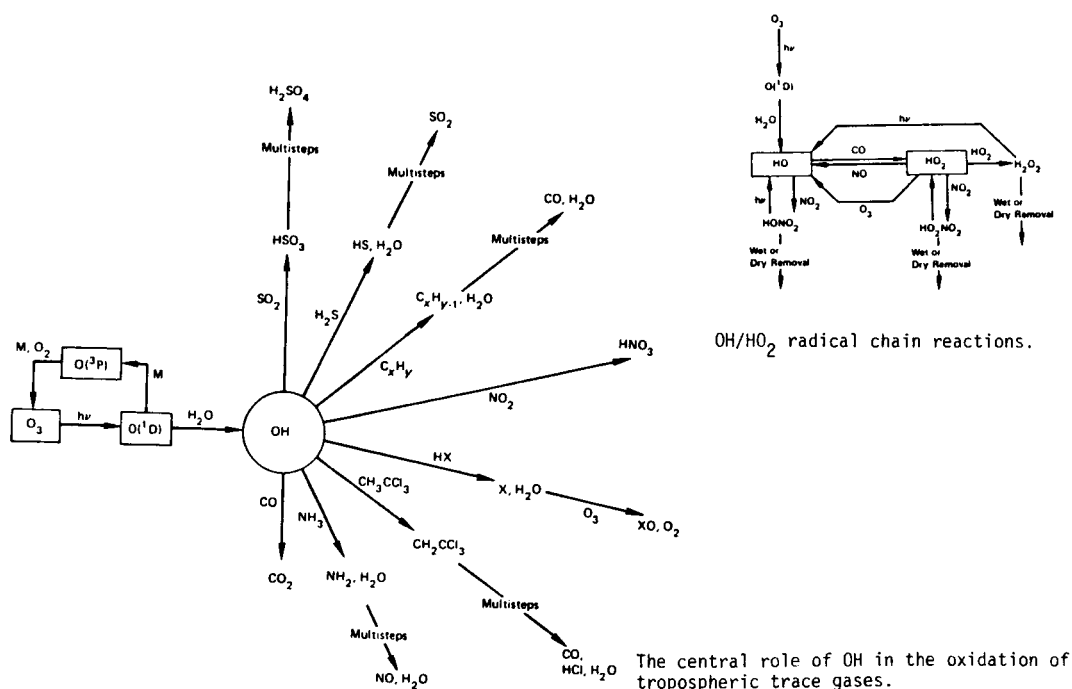
SPECIES	CONCENTRATION <sup>1</sup>	LIFETIME <sup>2</sup>	COMMENTS <sup>3</sup>
WATER VAPOR (H <sub>2</sub> O)	<< 1% - 4%	HYDROLOGICAL CYCLE: VAPOR FORM: ~10 DAYS DROPLET FORM: ~HOURS	EVAPORATION/CONDENSATION; LEADS TO OH PRODUCTION; GREENHOUSE SPECIES
MOLECULAR HYDROGEN (H <sub>2</sub> )	0.5 PPMV	$3.0 \times 10^7$ s ( $8.3 \times 10^3$ HR)	C, B
HYDROGEN PEROXIDE (H <sub>2</sub> O <sub>2</sub> )	$10^9$ CM <sup>-3</sup>	$1.2 \times 10^5$ s (33 HR)	C; WATER SOLUBLE
HYDROPEROXYL RADICAL (HO <sub>2</sub> )	$10^8$ CM <sup>-3</sup>	MINUTES	C
HYDROXYL RADICAL (OH)	$10^6$ CM <sup>-3</sup>	V.S.	C; PRINCIPAL OXIDIZER IN TROPOSPHERE
ATOMIC HYDROGEN (H)	$\sim 1$ CM <sup>-3</sup>	V.S.	C

<sup>1</sup>LEVINE, J.S. (EDITOR), 1985: THE PHOTOCHEMISTRY OF ATMOSPHERES: EARTH, THE OTHER PLANETS, AND COMETS, ACADEMIC PRESS, INC., PP. 6-7.

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## HYDROGEN SPECIES CHEMISTRY



FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICCERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.

# NITROGEN SPECIES

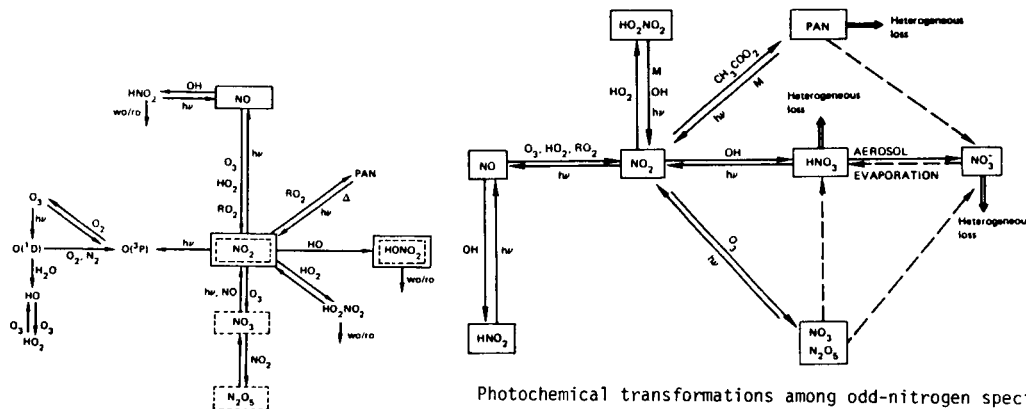
SPECIES	CONCENTRATION <sup>1</sup>	LIFETIME <sup>2</sup>	COMMENTS <sup>3</sup>
NITROUS OXIDE (N <sub>2</sub> O)	330 PPBV	V.L.	B, A; GREENHOUSE SPECIES; GLOBAL INCREASE (~0.22/YR)
AMMONIA (NH <sub>3</sub> )	0.1 - 10 PPBV	1.3 x 10 <sup>6</sup> s (360 HR)	B, A; WATER SOLUBLE; MAJOR GASEOUS BASE; GREENHOUSE SPECIES; AEROSOL PRODUCTION
NITRIC ACID (HNO <sub>3</sub> )	50 - 1000 PPTV	1.5 x 10 <sup>6</sup> s (420 HR)	C; WATER SOLUBLE; ACID RAIN; AEROSOL PRODUCTION
NITROGEN DIOXIDE (NO <sub>2</sub> )	10 - 300 PPTV	1.3 x 10 <sup>4</sup> s (3.6 HR)	C
NITRIC OXIDE (NO)	5 - 100 PPTV	1.1 x 10 <sup>2</sup> s (0.03 HR)	A, B, C, LIGHTNING
NITROGEN TRIOXIDE (NO <sub>3</sub> )	100 PPTV	-SECONDS	C
PEROXYACETYLNITRATE (CH <sub>3</sub> CO <sub>3</sub> NO <sub>2</sub> )	50 PPTV	DAYS TO YEARS	C
DINITROGEN PENTOXIDE (N <sub>2</sub> O <sub>5</sub> )	1 PPTV	-SECONDS	C
PERNITRIC ACID (HO <sub>2</sub> NO <sub>2</sub> )	0.5 PPTV	-DAYS	C; WATER SOLUBLE
NITROUS ACID (HNO <sub>2</sub> )	0.1 PPTV	-DAYS	C; WATER SOLUBLE

<sup>1</sup>LEVINE, J.S. (EDITOR), 1985: THE PHOTOCHEMISTRY OF ATMOSPHERES: EARTH, THE OTHER PLANETS, AND COMETS, ACADEMIC PRESS, INC., PP. 6-7.

<sup>2</sup>IBID, P. 44: V.S. = VERY SHORT (SECONDS OR LESS); V.L. = VERY LONG (DECADES OR LONGER)

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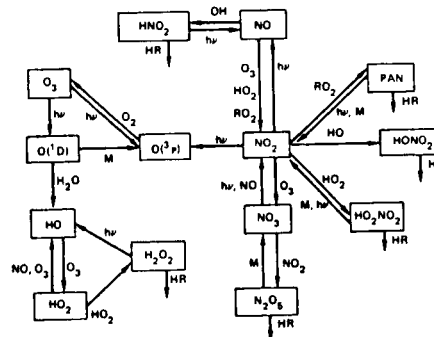
## NITROGEN SPECIES CHEMISTRY



Photochemical transformations among odd-nitrogen species.

Major atmospheric reactions of N<sub>x</sub>O<sub>y</sub> species: Solid line boxes indicate major daytime nitrogen species; broken line boxes indicate significant nighttime nitrogen species. It is still uncertain whether PAN is a major species in the free troposphere. The notation wo/ro denotes washout/rainout process.

FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.



Major atmospheric reactions of H<sub>x</sub>O<sub>y</sub>, N<sub>x</sub>O<sub>y</sub>, and O<sub>3</sub>, where M denotes N<sub>2</sub> and O<sub>2</sub>, HR refers to heterogeneous removal, and hv indicates radiation required.

# CARBON SPECIES

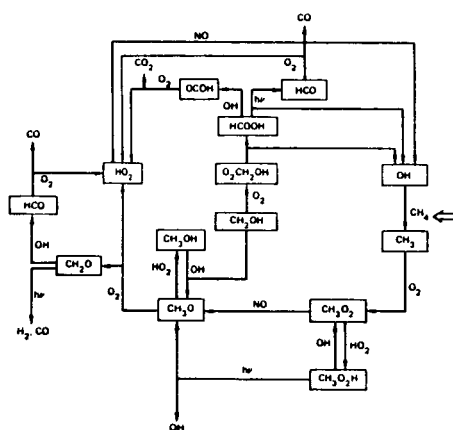
SPECIES	CONCENTRATION <sup>1</sup>	LIFETIME <sup>2</sup>	COMMENTS <sup>3</sup>
CARBON DIOXIDE (CO <sub>2</sub> )	0.034%	V.L.	A, B, VOLCANIC; GREENHOUSE SPECIES; GLOBAL INCREASE (-.2%/YR)
METHANE (CH <sub>4</sub> )	1.7 PPMV	2.5 x 10 <sup>7</sup> s (6.9 x 10 <sup>3</sup> HR)	B, A; GREENHOUSE SPECIES; GLOBAL INCREASE (-1.1%/YR)
CARBON MONOXIDE (CO)	70 - 200 PPBV (N.H.) 40 - 60 PPBV (S.H.)	7.1 x 10 <sup>5</sup> s (200 HR)	A, B, C; GLOBAL INCREASE (-1-2%/YR)
FORMALDEHYDE (H <sub>2</sub> CO)	0.1 PPBV	~HOURS	C; WATER SOLUBLE
METHYHYDROPEROXIDE (CH <sub>3</sub> OOH)	10 <sup>11</sup> CM <sup>-3</sup>	~DAYS	C; WATER SOLUBLE
METHYLPEROXYL RADICAL (CH <sub>3</sub> O <sub>2</sub> )	10 <sup>8</sup> CM <sup>-3</sup>	V.S.	C
METHYL RADICAL (CH <sub>3</sub> )	10 <sup>-1</sup> CM <sup>-3</sup>	V.S.	C

<sup>1</sup>LEVINE, J.S. (EDITOR), 1985: THE PHOTOCHEMISTRY OF ATMOSPHERES: EARTH, THE OTHER PLANETS, AND COMETS, ACADEMIC PRESS, INC., PP. 6-7.

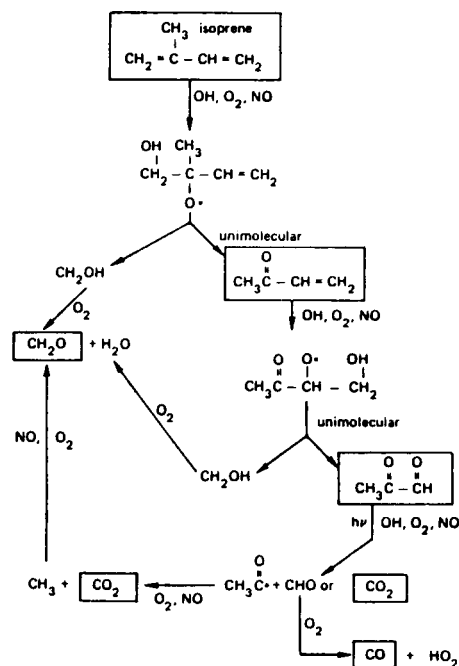
<sup>2</sup>IBID, P. 44: V.S. = VERY SHORT (SECONDS OR LESS); V.L. = VERY LONG (DECADES OR LONGER)

<sup>3</sup>A = ANTHROPOGENIC; B = BIOGENIC; C = CHEMICAL OR PHOTOCHEMICAL

## CARBON SPECIES CHEMISTRY



A possible tropospheric degradation scheme for CH<sub>3</sub>O<sub>2</sub> radicals, formed from CH<sub>4</sub>. The current lack of understanding of this chemistry defines one of the major uncertainties in the understanding of fast H<sub>x</sub>O<sub>y</sub> photochemistry.



A possible reaction scheme for isoprene oxidation in the presence of NO<sub>x</sub>.

FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.

81

# HALOGEN SPECIES

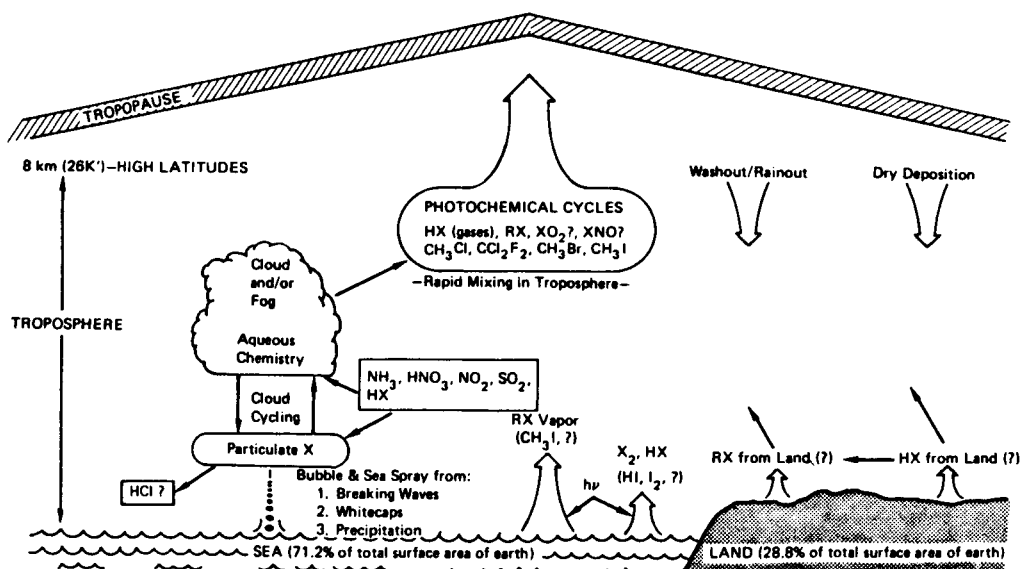
SPECIES	CONCENTRATION <sup>1</sup>	LIFETIME <sup>2</sup>	COMMENTS <sup>3</sup>
HYDROGEN CHLORIDE (HCl)	1 PPBV	$3.0 \times 10^5$ s (83 HR)	SEA SALT, VOLCANIC; WATER SOLUBLE
METHYL CHLORIDE (CH <sub>3</sub> Cl)	0.5 PPBV	$4.9 \times 10^6$ s ( $1.3 \times 10^3$ HR)	B, A
METHYL BROMIDE (CH <sub>3</sub> Br)	10 PPTV	~MONTHS	B, A
METHYL IODIDE (CH <sub>3</sub> I)	1 PPTV	~MONTHS	B, A

<sup>1</sup>LEVINE, J.-S. (EDITOR), 1985: THE PHOTOCHEMISTRY OF ATMOSPHERES: EARTH, THE OTHER PLANETS, AND COMETS, ACADEMIC PRESS, INC., PP. 6-7.

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<sup>3</sup>A = ANTHROPOGENIC; B = BIOGENIC; C = CHEMICAL OR PHOTOCHEMICAL

## HALOGEN SPECIES CHEMISTRY

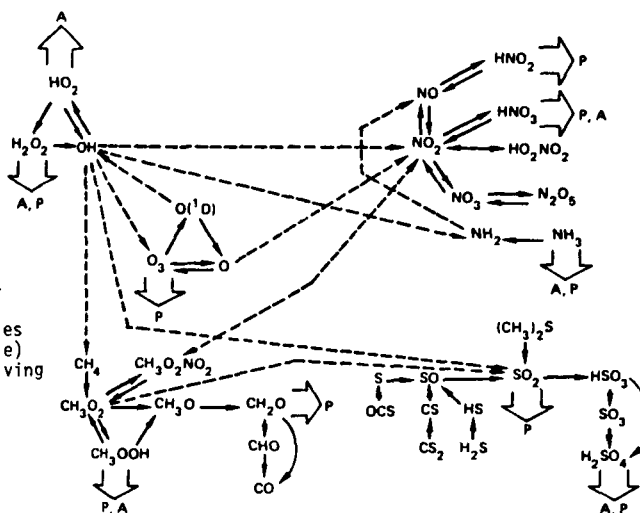


Schematic diagram to show processes and to exemplify key species in global tropospheric halogen cycles. X denotes F, Cl, Br, or I.

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Aerosols as an end product of atmospheric reactions. Major reaction pathways for gas-phase constituents are depicted by solid lines. Interactions between chemical families are indicated by dashed lines. Heavy (double) arrows show key heterogeneous pathways involving aerosols (A) and precipitation (P). Turco et al., 1982.\*

\*Turco, R. P., O. B. Toon, R. C. Whitten, R. G. Keese and P. Hamill, Importance of Heterogeneous Processes to Tropospheric Chemistry, Geophysical Monograph Series, vol. 26, D. R. Schryer, editor, American Geophysical Union, Washington, D.C., pp. 231-240, 1982.\*



FROM GLOBAL TROPOSPHERIC CHEMISTRY: A PLAN FOR ACTION, GLOBAL TROPOSPHERIC CHEMISTRY PANEL (R. A. DUCE, CHAIRMAN, AND R. CICERONE, VICE CHAIRMAN) PUBLISHED BY THE NATIONAL ACADEMY PRESS, WASHINGTON, DC, 1984.